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COATING MATERIAL COMPOSITIONS

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COATING MATERIAL COMPOSITIONS

[Toryo yo soseibutsu]

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Claims

1. The title coating material compositions contain (A) at least one polyol selected from polyesters and acrylic polyols, (B) functional fluoropolymers, (C) at least one binder selected from isocyanate compounds containing at least two functional groups, blocked isocyanate compounds and melamine compounds, (D) at least one inorganic organosol selected from [Periodic Table] Group II (Mg, Ca, Sr, Ba), Group III (B, Al, Ga, In, Tl, Sc, Y, La, Ac), Group IV (Si, Ge, Sn, Pb, Ti, Zr, Hf), Group V (As, Sb, Bi, V, Nb, Ta), Group VI (Te, Po, Cr, Mo, W), Group VII (At, Mn, Tc, Re), and Group VIII (Fe, Co, Ni, Ru, Rh, Pd, Os, Ir, Pt) metal oxide sols and salts of the metal oxide sols, siloxane, metallosiloxane, silazane, metallosilazane, and mixtures of these compounds, and (E) solvents.

2. Compositions described in Claim 1 that further contain (F) reaction-accelerating catalysts.

3. Compositions described in Claim 1 or 2 using, for inorganic organosols (D), the metal oxide sols selected from Group III (B, Al, Ga, In, Tl, Sc, Y, La, Ac), Group IV (Si, Ge, Sn, Pb, Ti, Zr, Hf), and Group V (As, Sb, Bi, V, Nb, Ta).

4. Compositions described in Claim 1 or 2 using, for inorganic organosols (D), the metal oxide sols selected from Si, Al, Sn, Ti, and Zr.

5. Compositions described in any of Claims 1-4, wherein the compounding ratios in weight percent of polyols (A), functional fluoropolymers (B), binders (C), and inorganic organosols (D) are (A): 10-80%, (B): 2-80%, (C): 2-50%, and (D): 5-60%.

6. Compositions described in any of Claims 1-4 that further contain pigments (G).

Detailed explanation of the invention

[0001]

Industrial application field

The present invention relates to coating material compositions that have excellent weatherability and abrasion resistance.

[0002]

The coating material compositions are useful as coating materials or coating agents for the surface of inorganic materials (steel sheets, aluminum sheets, glass, roofing tiles, slates, etc.) and organic materials (wood, paper, cellophane, plastic, coating films of organic coating materials, etc.).

[0003]

Examples of steel sheets coating targeted for include hot-rolled steel sheets, cold-rolled steel sheets, electrogalvanized steel sheets, hot-dipped galvanized steel sheets, , alloy-electroplated steel sheets, and those steel sheets having chemical conversion with chromic acid, phosphoric acid, etc., and in addition, tinplate sheet, tin-free steel sheets, stainless steel sheets, etc.

[0004]

They are especially useful in household appliances, construction, engineering works (buildings, houses, curtain walls, tanks, plants, marine structures, bridges, etc.), transportation vehicles (automobiles, trolley cars, airplanes, etc.).

[0005]

Prior art

To date, polyesters, acrylic polymers (including acrylic silicones), etc., or mixtures of at least two of these compounds have been used as polymers for coating materials with good workability.

[0006]

Problems to be solved by the invention

Polymers for coating materials with good workability that have been used up to now have problems of being easily damaged and easily worn away due to their low hardness. Additionally, because their weatherability (resistance to weather) is low, therefore, when they are exposed outdoors over a long period of time, they easily discolor, crack, chalk, etc., which are problems.

[0007]

Means to solve the problems

The present inventors carried out a detailed study to solve the above-mentioned problems and, as a result, they found that for substrates coated with compositions containing polyols, functional fluoropolymers, binders, inorganic organosols, and solvents, and then cured at room-temperature or baked, when fluoropolymers and inorganic organosols were introduced into the polyols through the binders, coatings were formed that had better weatherability and hardness than those of polyols, thereby they completed the present invention.

[0008]

Namely, the present invention provides coating material compositions containing (A) at least one polyol selected from polyesters and acrylic polyols, (B) functional fluoropolymers, (C) at least one binder selected from isocyanate compounds containing at least two functional groups, blocked isocyanate compounds and melamine compounds, (D) at least one inorganic organosol selected from [Periodic Table] Group II (Mg, Ca, Sr, Ba), Group III (B, Al, Ga, In, Tl, Sc, Y, La, Ac), Group IV (Si, Ge, Sn, Pb, Ti, Zr, Hf), Group V (As, Sb, Bi, V, Nb, Ta), Group VI (Te, Po, Cr, Mo, W), Group VII (At, Mn, Tc, Re), and Group VIII (Fe, Co, Ni, Ru, Rh, Pd, Os, Ir, Pt) metal oxide sols and salts of the metal oxide sols, siloxane, metallosiloxane, silazane, metallosilazane, and mixtures of these compounds, and (E) solvents.

[0009]

The polyols (A) used in the present invention include polyols, acrylic polyols, and mixtures thereof. The polyols can be obtained by condensation of (1) polybasic acids such as

phthalic acid, isophthalic acid, terephthalic acid, maleic acid, fumaric acid, succinic acid, adipic acid, sebacic acid, azelaic acid, trimellitic acid, etc., with (2) polyols such as ethylene glycol, diethylene glycol, propylene glycol, dipropylene glycol, 1,3-butanediol, 1,4-butanediol, 1,5-pantanediol, neopentyl glycol, hexamethylene glycol, decamethylene glycol, hydroquinone bis(hydroxyethyl ether), 2,2,4-trimethyl-1,3-pantanediol, hydrogenated bisphenol A, trimethylolethane, trimethylolpropane, hexanetriol, glycerol, pentaerythritol, tris(hydroxyethyl) isocyanurate, cyclohexanediol, cyclohexanedimethanol, xylylene glycol, etc., in the presence of excess hydroxyl groups using a known process. In this case it is possible to use jointly at least two kinds of acid or polyol, respectively. Castor oil and higher fatty acid may be used together to form the so-called oil-modified polyester polyols.

[0010]

Polyesters obtained by the above-mentioned combinations having molecular weight of about 500-300,000, preferably about 2,000-100,000 and hydroxyl value of about 5-300, preferably about 10-200 are used preferably. The acrylic polyols can be obtained by copolymerization of one hydroxyl group-containing monomers obtained from the reaction of 2-hydroxyethyl acrylate, 2-hydroxypropyl acrylate, 2-hydroxyethyl methacrylate, 2-hydroxypropyl methacrylate, allyl alcohol, cinnamyl alcohol, crotonyl alcohol, or unsaturated carboxylic acids such as acrylic acid, methacrylic acid, maleic acid, fumaric acid, crotonic acid, itaconic acid, etc., with ethylene glycol, ethylene oxide, propylene glycol, propylene oxide, butylenes glycol, 1,4-cyclohexanedimethanol, phenyl glycidyl ether, glycidyl decanoate, etc., with (2) acrylic acid esters such as methyl acrylate, ethyl acrylate, n-propyl acrylate, isopropyl acrylate, n-butyl acrylate, tert-butyl acrylate, 2-ethylhexyl acrylate, etc.; methacrylic acid esters such as methyl methacrylate, ethyl methacrylate, n-propyl methacrylate, isopropyl methacrylate, n-butyl methacrylate, tert-butyl methacrylate, 2-ethylhexyl methacrylate, etc.; styrene-type monomers such as styrene, vinyltoluene, α -methylstyrene, etc.; and other copolymerizable α,β -ethylenic unsaturated monomers such as acrylic acid, methacrylic acid, vinyl acetate, vinyl propionate, acrylonitrile, vinyl stearate, allyl acetate, diallyl adipate, dimethyl itaconate, diethyl maleate, vinyl chloride, vinylidene chloride, ethylene, glycidyl methacrylate, N-methylolacrylamide, N-butoxymethylacrylamide, acrylamide, diacetone acrylamide, etc. Two or more of these copolymerizable α,β -ethylenic unsaturated monomers may be copolymerized together.

[0011]

For the acrylic polyols obtained by the combinations of the above-mentioned starting materials, those acrylic polyols having molecular weight about 1,000-500,000, preferably about 5,000-100,000 and hydroxyl value about 5-300, preferably about 10-200 are preferably used.

[0012]

Examples of polyesters which can be used as commercially available products include Desmophen 670, 680, and 850 (Sumitomo Bayer Urethane Co., Ltd.); Burnock D161, D6-439, and D220 (Dainippon Ink and Chemicals, Inc.); Nippollan 136 (Nippon Polyurethane Industry Co., Ltd.), etc. Examples of commercially available acrylic polyols include Arothane UW2818 (Nippon Shokubai Kagaku Kogyo Co., Ltd.); Acrydic A801, A811, and A808 (Dainippon Ink and Chemicals, Inc.); Hitalloid 2462A and 2405 (Hitachi Chemical Co., Ltd.), Desmophen A160, A165, and A260 (Sumitomo Bayer Urethane Co., Ltd.), etc.

[0013]

Examples of functional groups in functional fluoropolymers used in the present invention include hydroxyl group, carboxyl group, amino group, etc. However, hydroxyl group is especially preferable. Thus, for the functional fluoropolymers (B), binary copolymers from fluorine-containing vinyl monomers and functional group-containing vinyl monomers and ternary copolymers from fluorine-containing vinyl monomers and functional group-containing vinyl monomers and other copolymerizable vinyl monomers can be used.

[0014]

Examples of the fluorine-containing vinyl monomers include vinyl fluoride, vinylidene fluoride, trifluoroethylene, tetrafluoroethylene, bromotrifluoroethylene, chlorotrifluoroethylene, pentafluoropropylene, hexafluoropropylene, (per)fluoroalkyl trifluorovinyl ether, etc. Especially those compounds having large proportion of fluorine in the molecules are preferable.

[0015]

Among the functional group-containing vinyl monomers, examples of hydroxyl group-containing vinyl monomers include hydroxyalkyl vinyl ethers such as hydroxyethyl vinyl ether, hydroxypropyl vinyl ether, and hydroxybutyl vinyl ether; and hydroxyalkyl (meth)acrylates such as 2-hydroxyethyl (meth)acrylate, 2-hydroxypropyl (meth)acrylate, and diethylene glycol (meth)acrylate. Examples of carboxyl group-containing vinyl monomers include acrylic acid, methacrylic acid, maleic acid, maleic anhydride, itaconic acid, itaconic anhydride, fumaric acid, etc. Examples of amino group-containing vinyl monomers include

dimethylaminoethyl vinyl ether, dimethylaminopropyl vinyl ether,
N,N-dimethylaminopropyl(meth)acrylamide, dimethylaminoethyl (meth)acrylate, etc.

[0016]

Examples of other copolymerizable vinyl monomers include alkyl vinyl ethers such as ethyl vinyl ether, n-butyl vinyl ether, isobutyl vinyl ether, and cyclohexyl vinyl ether; vinyl esters of linear or branched-chain aliphatic carboxylic acids such as vinyl acetate, vinyl propionate, vinyl butyrate, vinyl pivalate, vinyl caproate, vinyl caprate, vinyl caprylate, etc.; alicyclic carboxylic acid vinyl esters such as cyclohexanecarboxylic acid vinyl ester; aromatic carboxylic acid vinyl esters such as vinyl benzoate, vinyl p-tert-butylbenzoate, vinyl salicylate, etc.; epoxy group-containing vinyl monomers such as glycidyl vinyl ether, glycidyl methacrylate, etc.; carboxyl group-containing vinyl monomers such as acrylic acid, methacrylic acid, maleic acid, maleic anhydride, itaconic acid, itaconic anhydride, fumaric acid, monoethyl maleate, monobutyl maleate, monobutyl fumarate, monobutyl itaconate, monovinyl adipate, monovinyl sebacate, etc.; amino group-containing vinyl monomers such as dimethylaminoethyl vinyl ether, dimethylaminopropyl vinyl ether, N,N-dimethylaminopropyl(meth)acrylamide, and dimethylaminoethyl (meth)acrylate; halogen (excluding fluorine)-containing vinyl monomers such as vinyl chloride, vinylidene chloride, etc.; aromatic vinyl monomers such as styrene, α -methylstyrene, vinyltoluene, and vinylpyridine; (meth)acrylic acid esters such as methyl (meth)acrylate, ethyl(meth)acrylate, butyl (meth)acrylate, 2-ethylhexyl (meth)acrylate, lauryl (meth)acrylate, cyclohexyl (meth)acrylate, and β -hydroxyethyl (meth)acrylate; (meth)acrylonitrile, (meth)acrylamide, N-methylol(meth)acrylamide, N-butoxymethyl(meth)acrylamide, maleic acid diamide, etc.

[0017]

At least two each of fluorine-containing vinyl monomers, functional group-containing vinyl monomers, and copolymerizable vinyl monomers may be used together.

[0018]

Examples of commercially available fluoropolymers which can be used include Lumiflon (Asahi Glass Co., Ltd.), Cefral Coat (Central Glass Co., Ltd.), Fluonate (Dainippon Ink and Chemicals, Inc.), Kynar (Pennwalt Corp., USA), Neoflon (Daikin Industries, Ltd.), Birahru [transliteration] (Hoechst AG, West Germany), etc.

[0019]

Binder (C) can be selected from isocyanate compounds having at least two functional groups, blocked isocyanate compounds, and melamine compounds. One or a mixture of at least two of these compounds may be added to the functional fluoropolymers.

[0020]

Aliphatic, alicyclic, or aromatic isocyanate compounds or other polycyanate compounds or their modified compounds may be used as the isocyanate compounds. However, for the weatherability of the coatings (paint films), especially for preventing yellowing, it is preferable to use non-yellowing modified polyisocyanate compounds that do not contain isocyanate groups directly bonded to aromatic rings. Examples of non-yellowing polyisocyanate compounds include hexamethylene diisocyanate, 1,4-cyclohexanebis(methyl isocyanate), methylenebis(cyclohexyl isocyanate), cyclohexylmethane diisocyanate, isophorone diisocyanate, 2-isocyanatoethyl 2,6-diisocyanatohexanoate, methyl 2,6-diisocyanatocaproate, dimethyl diisocyanate, dianisidine diisocyanate, cyanuric acid, isocyanuric acid, and their modified products. The modified products include trimer-type, dimer-type, prepolymer-type, biuret-type, urea-type and other modified products. Particularly suitable modified products such as trimer-type, prepolymer-type such as trimethylolpropane-modified products, and biuret-type modified products. If circumstances require, instead of these non-yellowing polycyanate compounds, either (1) aromatic yellowing polyisocyanate compounds or (2) both non-yellowing polycyanate compounds and aromatic yellowing polyisocyanate compounds, can be used.

[0021]

In the present specification, the blocked isocyanate compounds means those blocked isocyanate compounds which can be de-blocked to produce isocyanate groups under the following curing conditions and they can be prepared by blocking reaction of the above-mentioned polyisocyanate compounds. Suitable blocking agents may be selected according to uses and curing conditions. For example lactam-type blocking agents such as ϵ -caprolactam; oxime-type blocking agents such as acetoxime, methyl ethyl ketoxime, methyl isoamyl ketoxime, methyl isobutyl ketoxime, and other oxime-type blocking agents; phenol-type blocking agents such as phenol, cresol, catechol, nitrophenol, and other phenol-type blocking agents; alcohol-type blocking agents such as isopropanol, trimethylolpropane, and other alcohol-type blocking agents; and other active methylene-type blocking agents such as malonic acid ester, acetoacetic acid ester, etc., may be selected; however, it is not limited to the compounds mentioned here. Preferable blocking agents include lactam-type and oxime-type blocking agents.

[0022]

Generally speaking, in the case of using as compositions for a room-temperature-curable hard coating, it is preferable to contain isocyanate compounds, and in the case of using as compositions for a baked hard coating it is preferable to contain blocked isocyanate compounds. However, a combination of at least two binders may be used as well.

[0023]

Examples of commercially available isocyanate-type binders include Coronate 2515 (Nippon Polyurethane Industry Co., Ltd.), Desmodur BL3175 (Sumitomo Bayer Urethane Co., Ltd.), etc.

[0024]

Examples of melamine compounds include dimethylolmelamine, trimethylolmelamine, tetramethylolmelamine, pentamethylolmelamine, hexamethylolmelamine, isobutyl ether-type melamine, n-butyl ether-type melamine, butylated benzoguanamine, etc.

[0025]

For inorganic organosols (D), at least one inorganic organosol selected from [Periodic Table] Group II (Mg, Ca, Sr, Ba), Group III (B, Al, Ga, In, Tl, Sc, Y, La, Ac), Group IV (Si, Ge, Sn, Pb, Ti, Zr, Hf), Group V (As, Sb, Bi, V, Nb, Ta), Group VI (Te, Po, Cr, Mo, W), Group VII (At, Mn, Tc, Re), and Group VIII (Fe, Co, Ni, Ru, Rh, Pd, Os, Ir, Pt) metal oxide sols and salts of the metal oxide sols, siloxane, metallosiloxane, silazane, metallosilazane, and mixtures of these compounds may be used. Preferable inorganic organosols (D) include metal oxide sols selected from Group III (B, Al, Ga, In, Tl, Sc, Y, La, Ac), Group IV (Si, Ge, Sn, Pb, Ti, Zr, Hf), and Group V (As, Sb, Bi, V, Nb, Ta), and particularly preferable metal oxide sols are those in which the metals are selected from Si, Al, Sn, Ti, and Zr. The salts of metal oxide sols means the salts formed from metal oxide sols and cations (Li^+ , K^+ , Na^+ , Mg^{2+} , Zn^{2+} , Al^{3+} , Ca^{2+} , etc.) or anions (S^{2-} , PO_4^{3-} , SO_4^{2-} , CH_3COO^- , NO_3^- , Cl^- , F^- , etc.)

[0026]

The compounding ratios [in weight percent] of polyols (A), functional fluoropolymers (B), binders (C), and inorganic organosols (D) are preferably (A): 10-80%, (B): 2-80%, (C): 2-50%, and (D): 5-60%, more preferably (A): 20-60%, (B): 5-60%, (C): 5-30%, and (D): 10-50%.

[0027]

For the solvents (E), suitable amount of solvents that can dissolve each above-mentioned compounding component can be used. Particularly suitable are organic solvents that have high solubility for functional fluoropolymers (B) and inorganic organosols (D), such as xylene, toluene, other aromatic hydrocarbon solvents; butyl acetate, other ester-type solvents; methyl isobutyl ketone, cyclohexanone, other ketone-type solvents; ethyl cellosolve, other glycol ether-type solvents, Carbitol acetate, other diethylene glycol ester-type solvents; and various kinds of thinners. Of course they are not limited to those mentioned here; thus, other hydrocarbons, halogenated hydrocarbons, alcohols, phenols, acetals, esters, ethers, ketones and other solvents can be used. These solvents can be suitably selected and used after taking into consideration the types and conditions of substrates, evaporation rate, working environment, and other conditions; however, particularly from the viewpoint of easily obtained good coatings the use of ketone-type solvents is preferable.

[0028]

From the viewpoint of acceleration of curing and improvement of workability, the addition of reaction-promoting catalysts (F) to the compositions of the present invention is preferable.

[0029]

For the catalysts (F), basic or acidic curing catalysts can be used. Examples of the basic catalysts include dibutyltin dilaurate, dibutyltin diacetate, stannous octanoate, other organotin compounds, methylimidazole, acridine, triethylamine, hexadecyltrimethylammonium stearate, and other amine-type catalysts. Examples of the acidic catalysts include methanesulfonic acid, dodecylbenzenesulfonic acid, and toluenesulfonic acid, which are organic sulfonic acids. For the catalysts (F), at least one of these basic catalysts or acidic catalysts may be used. However, co-catalysts may be jointly used as well.

[0030]

In addition to the above-mentioned (A)-(E) essential components and catalysts (F), depending on the objective, other components may be compounded into the compositions of the present invention for coating materials. For example when they are used as coating materials, pigments (G) may be added if necessary.

[0031]

Examples of the pigments (G) include inorganic pigments such as chrome yellow, molybdate orange, Prussian blue, cadmium-based pigments, titanium white, composite oxide pigments, transparent iron oxides and organic pigments such as cyclic quality pigments, soluble azo dyes, insoluble azo dyes, copper phthalocyanine pigments, dyeing pigments, pigment intermediates, etc.

[0032]

Coloring agent, thixotropy agent, filler, thickener, leveling agent, antifoaming agent, stabilizer, and other additives may be compounded as well.

[0033]

The compositions of the present invention for coating materials can be manufactured by mixing the above-mentioned components using a known process.

[0034]

Using well-known methods such as spray coating, brush coating, roll coating, etc., substrates can be coated with the compositions of the present invention for coating materials and then allow to stand at room temperature for 1-10 days to cure, or baked at 60-250°C for 1-30 minutes to cure.

[0035]

Effect of the invention

The compositions of the present invention for coating materials have more excellent weatherability and improved surface hardness than those of conventional polyester and acrylic polymer coating materials having good workability, therefore they have excellent abrasion resistance and antifouling property as well, so that they can be used in precoated metals for external panels for buildings, household appliances, etc., and in factories, constructions in marine structures, construction works, automobiles, electric vehicles, etc.

[0036]

Application examples

Application Examples 1-9 and Comparative Examples 1-2

Coating material compositions were manufactured using the following starting materials and compounding ratios shown in Table 1. The amount compounded was indicated by weight (g). A 0.3 mm-thick stainless steel sheet (SUS 304) was coated with the above-mentioned

coating material compositions so the thickness of the coating material compositions after baking was about 20 μ . It was baked at 160°C for 20 minutes for curing. The physical properties of the coating (films) are shown in Table 1.

[0037]

(1) Polyol

- O Acrylic polyol: Arothane UW2818 (nonvolatile matter: 60 wt%)
- O Polyester: Desmophen 850 (nonvolatile matter: 100 wt%)

(2) Functional fluoropolymer

- O Cefral Coat A-101B (nonvolatile matter: 55 wt%)
- O Lumiflon LF 200 (nonvolatile matter: 60 wt%)

(3) Isocyanate compound

- Coronate 2515 (nonvolatile matter: 80 wt%)

(4) Organosol

- O Organotitania sol (C-1) (nonvolatile matter: 30 wt%)
- O Organosilica sol (C-2) (nonvolatile matter: 30 wt%)

(5) Solvent

- Cyclohexane-type solvent

(6) Curing catalyst

- Dibutyltin dilaurate

Method for testing physical properties of coating (film)

- (1) Gloss: 60° reflectance prescribed by JIS K 5400
- (2) Hardness: Pencil hardness prescribed by JIS K 5400
- (3) Bending: Bending tester ϕ 2 mm

[0038]

"○" means the tests were passed.

[0039]

- (4) Xylene: Xylene wipe-off test

[0040]

(5) Weatherability: Sunshine weatherometer accelerated weather resistance test 3000 hours

○ Retention of initial gloss: 90% or greater

○ Retention of initial gloss: 85% or greater

[0041]

Table 1

①	■ ■	② 実験例					③ 金型樹	② 実験例				③ 金型樹
		1	2	3	4	5		1	6	7	8	
④	■ ■	アロタン UW218 / セフラルコート A-101B 80/20	5	アロタン UW218 100	6	デスマーフェン 850 / ルミフロン LF 100 20/20	7	デスマーフェン 850 30/30	8			
9	イリシアネット	33	34	31	33	34	32	33	34	31	34	33
10	オルガノソル C-I C-II	50	25	100	35	75		48	75		59	75
11	溶 剤	100	15	30	30	15	33	100	15	200	15	10
12	触媒	8.5	8.5	8.5	8.5	8.5	8.5	6.5	3.5	8.5	8.5	8.5
13	物性											
14	光 漆	85	85	82	81	84	85	84	83	84	81	82
15	硬 度	55	55	55	55	55	55	55	55	55	55	55
16	折り曲げ	○	○	○	○	○	○	○	○	○	○	○
17	キシレン	200	230	250	230	250	230	200	240	260	250	200
18	耐 烟	◎	◎	○	◎	◎	○	◎	◎	◎	◎	○

Key:

- 1 Compound
- 2 Application Example
- 3 Comparative Example
- 4 Polymer
- 5 Arothane UW218/Cefral Coat A-101B 80/20
- 6 Arothane UW218 100
- 7 Desmophen 850/Lumiflon LF 200
- 8 Desmophen 850 100
- 9 Isocyanate
- 10 Organosol
- 11 Solvent
- 12 Catalyst
- 13 Physical properties of coating (film)
- 14 Gloss
- 15 Hardness
- 16 Bending

17 Xylene
18 Weatherability